

## SPECTRAL RADIOMETRY AND TROPOSPHERIC AEROSOLS: REPORT OF PANEL

R. S. Fraser\* (Chairman)  
M. Griggs, A. A. Lacis, and L. R. McMaster\*

## INTRODUCTION

The term aerosols, as used here, refers to the haze, smoke, and dust that appear in the troposphere. The term does not refer to the hydrometeors in cumulus and stratus clouds but does include the sulfuric acid-water droplets which are assumed to predominate in the stratospheric aerosol layer. The aerosol properties that have been measured from satellites and those which can be made in the near term (up to 1992) will be reviewed. The capabilities that will exist in the years 1992-2000, with implementation of EOS, will be discussed next. Finally, a few words will be said concerning the potential for aerosol measurements for the decade after 2000.

## CURRENT CAPABILITIES

Two satellite methods have been, and are expected to be, available for measuring aerosol properties up to 1992. The first depends on receiving scattered sunlight from a column of the Earth-atmosphere system extending to the ground, and the other method utilizes solar occultation observations of the Earth's limb. The first method consists of measuring the intensity of scattered sunlight in the visible and near-infrared spectrum. The intensity is linearly proportional to aerosol properties such as mass, surface area, and number. Furthermore, those particles that scatter light most strongly are those with diameters approximately equal to the wavelength. Particles that are an order of magnitude smaller, or larger, are ineffective optically--the smaller ones because of their small cross sections and the larger particles because there are not enough of them. A few exceptions can be made, however. If there are enough large particles, such as is the case for Saharan dust, then they are visible at wavelengths much smaller than their particle size. With the exception of dust and, possibly, aerosols derived from the ocean, the available sensors measure properties in the size range between 0.1 and 1  $\mu\text{m}$ .

Several sensors are expected to be available in the near term. These include NOAA's polar orbiters carrying the AVHRR and their geostationary satellites carrying the VISSR. The nadir spatial resolution of these sensors is 1 km. AVHRR scans the entire Earth once each day. VISSR scans the Earth within 60° of the subsatellite point every half hour. The prime purpose of each sensor is to take cloud pictures. As a result, neither sensor is calibrated after launch. Post-launch calibration methods (Griggs, 1984; Fraser and Kaufman, 1986) have been developed, however. Other countries may also continue to deploy geostationary satellites with VISSR-type sensors. Research data will be available from observations made from Landsat and other U.S. and foreign sensors.

Nadir-viewing satellite sensors measure the total vertical burden of the aerosol properties. As a result, validation data are difficult to acquire and do not exist

---

\*Prepared panel report.

generally. The one exception is the aerosol optical thickness. It is linearly proportional to the intensity of the scattered light, at least for the common amounts of aerosol, just as for other aerosol properties previously mentioned. The accuracy of these optical thickness measurements indicates the accuracy of the mass, area, and number measurements also.

The optical thickness measurements are validated by values derived from the solar transmission to the ground. The accuracy of the satellite values is highest when observations are made over a uniform, dark surface such as the ocean. The first extensive measurements were made by Griggs (1984) over many oceanic regions. He compared satellite values with values derived from ground-based measurements of solar transmission (fig. 16). The standard error of estimate is 0.05. Measurements over land are more difficult because of variable surface reflectance, which is about the same or more than aerosol reflectance. Nevertheless, the surface reflectance can be monitored on days with small aerosol thickness. Values of optical thickness measured from a satellite at several locations are compared with ground-based values in figure 17. The good agreement results in the square of the correlation coefficient  $r^2 = 0.89$ .

Previous values of optical thickness measured from a nadir-viewing satellite were based on measurements in one spectral band. If two or more bands are used, then information about the distribution of physical properties, with respect to size, can be derived. The intensity of light scattered by aerosols relative to the solar spectral irradiance, or aerosol reflectance, depends on the aerosol size distribution. If the particles are much smaller than the wavelength, the aerosol reflectance varies as  $\lambda^4$ ; if the particles are larger than the wavelength, the reflectance is independent of wavelength. Usually the reflectance varies as  $\lambda^{-4}$  to  $\lambda^{-2}$ . The wavelength dependence of an aerosol size parameter has been derived from satellite measurements, but the results have been validated with only a few independent observations (Griggs, 1984; and Fraser and Kaufman, 1985).

An example of deriving the aerosol albedo of single scattering from Landsat observations of Washington, D.C., during August 1982, is given in figure 18. The reflectances of dark pixels increased from 0.03 to 0.13 with increasing turbidity. The intensity of light from pixels with surface reflectance of 0.18 changed only slightly. If one assumes that the aerosol particles are small and that their absorption cross section is much greater than their scattering cross section, then their albedo of single scattering is 0.9. This value was lower than the satellite value of 0.96 associated with hazes over the eastern U.S. during the summer. In the former case, however, smoke from forest fires over northwestern Canada had drifted over Washington, indicating the presence of soot.

The only remotely sensed clue concerning the chemical composition of the aerosols is the increased size of aerosol particles caused by condensation of water vapor. Then the decrease in optical thickness with respect to wavelength becomes less and, at the same time, the optical thickness increases.

Before global maps of aerosol properties can be derived from nadir-viewing satellite observations, efficient algorithms must be developed. The algorithms have to account for calibration, surface reflectance, and table lookups relating the aerosol properties of models with the intensity of scattered light. The models will depend on the geographical region and season of the year. Extensive tables have to be prepared as a function of solar zenith angle, viewing angles, and wavelength.

Before such nadir-viewing satellite measurements of aerosol properties are accepted, they need to be validated with independent data. Valuable experiments can be made at the surface such as the relation between the mass of sulfate and the scattering coefficient of light (Pierson et al., 1980) or the mass of water condensed on dry particles as a function of humidity. Nevertheless, such experiments are not sufficient since the satellite measures the average properties along a line of sight and the properties vary along the line of sight. Measured vertical profiles are desirable, and they can be obtained by aircraft. Since the aircraft experiments are difficult and expensive, remote measurements from the ground may have to be relied on if several independent measurements yield the same result. For example, measurements of solar transmission and skylight intensity and polarization in many spectral bands may all show the same distribution of aerosol mass with respect to size, giving greater confidence in these satellite data.

The second method of measuring aerosol optical properties utilizes solar occultation of the Earth's limb. This technique has been successfully employed from space with the Stratospheric Aerosol Monitor II (SAM II) flown on Nimbus 7, the Stratospheric Aerosol and Gas Experiment (SAGE) flown on the Application Explorer Mission 2 (AEM-2), and SAGE II flown on the ERBS. In each case, the instrument measures the attenuation of solar radiation through the Earth's limb during spacecraft sunrise and sunset. Such a measurement is essentially self-calibrating, ratioing the attenuated signal through the atmosphere to the unattenuated signal outside the atmosphere to obtain atmospheric transmission profiles.

These data are then inverted to retrieve vertical profiles of aerosol extinction coefficient from cloud tops to approximately 35 km, with a vertical resolution of 1 km and an accuracy of better than 10 percent at the peak of the stratospheric aerosol layer (McCormick et al., 1979; and Mauldin et al., 1985). In cloud-free regions, these profiles can extend down to the Earth surface and do, in fact, reach altitudes of 8 km or lower 50 percent of the time (Woodbury and McCormick, 1983), allowing studies of the upper troposphere as well as the stratosphere. Since these instruments measure the average extinction through the Earth's limb along the spacecraft to Sun view angle, the horizontal resolution of these profiles is  $1 \text{ km} \times 200 \text{ km}$ . Successive vertical profiles are separated by approximately  $24^\circ$  longitude as the observations sweep through the latitude extremes in 3 to 4 weeks, depending on orbit inclination.

SAM II, launched in 1978 and still operating today, employs a single spectral channel for measurement of aerosol extinction at one wavelength. The Nimbus 7 Sun-synchronous orbit provides geographic coverage from  $64^\circ$  to  $80^\circ$  latitude in both the Northern and Southern Hemispheres. Thus, SAM II is accumulating a relatively large data base on polar aerosols (McCormick, 1981; and McCormick, 1983). SAGE I, launched in 1979, employed four spectral channels for measurement of ozone and nitrogen dioxide in addition to aerosol extinction at two wavelengths. The AEM-2  $56^\circ$  inclination orbit provided geographic coverage from  $79^\circ$  south to  $79^\circ$  north latitude. Thirty-three months of SAGE data were collected before the AEM spacecraft power subsystem developed problems, preventing further instrument operation. However, the almost 3 years of data have been used with the SAM II data set to develop a global upper tropospheric and stratospheric aerosol climatology (McCormick, 1983; McCormick, 1985; Kent and McCormick, 1984).

SAGE II, launched in 1984 and still operating today, employs seven spectral channels for measurement of ozone, nitrogen dioxide, and water vapor, in addition to aerosol extinction at four wavelengths. These data are currently being validated through comparisons with correlative observations such as ground-based and airborne

aerosol lidar backscatter data and balloon-borne in situ particle counters. An example of the preliminary SAGE II aerosol extinction data shown in figure 19 will then be used to study aerosol size and distribution and other aerosol optical properties (Yue et al., 1985).

The ERBS/SAGE II 57° inclination orbit provides geographic coverage from 80° south to 80° north latitude, repeating global coverage every 3 to 4 weeks. Since there are no expendables to be used up, SAGE II could continue to collect data into the early 1990's, providing an important data base for aerosols from the mid troposphere up to 35 km.

#### PERIOD: 1992-2000

Since the final stage of some important chemical cycles results in the formation of aerosols, the strength of aerosol sources and sinks and their locations are important for understanding atmospheric chemistry. To compute aerosol transport, convergence, and divergence, the aerosol vertical profile and the wind vector profile are required. Lidar can provide the vertical profile of the scattering coefficient. With more than one spectral band, the lidar also gives information about the size dependence of aerosol properties. The lidar data will be relatively infrequent, but the other EOS sensors will provide data on the total vertical burden of aerosol properties on a very small scale. Wind information will be available from standard meteorological data.

Abundant new data rich in spectral information will be available from EOS sensors. A challenge exists to develop algorithms for extracting the information. It is very important to include plans for validating the derived products. Another improvement that EOS will provide is good calibration. Current satellite sensors that measure the intensity of scattered sunlight are either not calibrated or poorly calibrated after launch. The measured reflectance should be accurate within 0.5 percent to measure aerosol properties with accuracy of 10 percent.

The chemical composition of aerosols is exceedingly difficult to measure from satellites. Remote measurements of composition of aerosols in the Earth's atmosphere from the surface of the Earth do not seem to have been made. Laboratory experiments have commenced, however, to identify aerosol species by means of laser backscatter (Goetz et al., 1985). Aerosols in the lower troposphere are screened from satellite observations in much of the spectrum by trace gas absorption. Electronic transitions at energies greater than 3 eV are unobservable because of ozone absorption. Aerosol absorption in the visible and near infrared is dominated by grey absorption. Aerosol vibration bands occur in the 1 to 3  $\mu\text{m}$  window (Bohren and Huffman, 1983) and also in the 8 to 13  $\mu\text{m}$  window (Goetz et al., 1985; C. Prabhakara, NASA Goddard, private communication, 1985). Aerosol spectra will be difficult to measure because of similar spectra from the Earth's surface, numerous absorption lines of water vapor and other trace gases, the small optical thickness of aerosols, and the optical constants are not known for aerosols with mixed composition.

C. Prabhakara (NASA Goddard, private communication, 1985) attempted to extract evidence for the strong silicate band at  $1100\text{ cm}^{-1}$  in the Saharan dust as it swept out over the Atlantic Ocean. He used the measurements from the infrared interferometer spectrometer (IRIS) with a spectral resolution of  $2.8\text{ cm}^{-1}$  and flown on Nimbus 4. He did not observe the strong Si-O emission near  $1100\text{ cm}^{-1}$ , but rather grey emission. Bohren and Huffman (1983) show that such grey emission is expected for dispersed sizes and nonspherical shapes, as are likely for the aeolian dust (Martin and Barber,

1985). This is the only attempt that I know about to identify the chemical composition of aerosols from satellite observations.

To show the difficulty in deriving aerosol spectral information from thermal emission, consider the important aerosol ammonium sulfate. Take as an example measurement of the difference in energy at a satellite from two regions where the difference in ammonium sulfate concentration is  $10 \mu\text{g}/\text{m}^3$ , as might occur between rural and urban areas (Remsberg, 1978). In order to estimate a relatively strong effect, assume that the aerosol is uniformly mixed in a layer 1500 m deep. Then the total mass of aerosol is  $0.015 \text{ g}/\text{m}^2$ . The absorption and scattering optical thicknesses for a typical accumulation mode, where sulfates are concentrated, are given in the table below for the minimum, intermediate, and maximum values of extinction in a strong band in an atmospheric window.

A linearized expression for the apparent temperature difference ( $\Delta T$ ) between the rural and urban areas caused by only the ammonium sulfate is

$$\Delta T = \tau_a [T_A - \epsilon_s T_s]$$

where  $\tau_a$  is the vertical aerosol absorption optical thickness,  $T_A$  is the temperature of the aerosol, and  $\epsilon_s$  and  $T_s$  are the emissivity and temperature of the surface, respectively. The maximum  $\tau_a = 0.0068$  in the following table, and if the temperature difference between the aerosol and surface emitting as a blackbody is 10K, then  $\Delta T = -0.07\text{K}$ . Such a small value would seem to be undetectable because of temperature fluctuations caused by the atmosphere and surface, plus radiometer noise.

#### OPTICAL PARAMETERS OF AMMONIUM SULFATE FOR A STRONG ABSORPTION BAND IN AN ATMOSPHERIC WINDOW\*

Wavelength ( $\mu\text{m}$ )	Index of Refraction		$C_{\text{ext}} \div \text{mass}$ ( $\text{m}^2\text{g}^{-1}$ )	Albedo of Single Scattering	Optical Absorption	Thickness Scattering
	Real	Imaginary				
8.31	1.2183	0.0514	0.042	0.0056	0.0006	$0.4 \times 10^{-5}$
8.71	1.0227	0.2759	0.234	0.0013	0.0035	$0.5 \times 10^{-5}$
9.07	1.3909	0.7055	0.458	0.0045	0.0068	$3.1 \times 10^{-5}$

\*The index of refraction is given by Remsberg (1978) for a relative humidity of 41 percent. The logarithm of the sizes is distributed normally with the geometric mean radius =  $0.027 \mu\text{m}$  and the geometric standard deviation = 2.16. The mass density is  $10 \mu\text{g}/\text{m}^3$ , uniformly mixed in layer 1500 m deep.

Lidar detection of the ammonium sulfate aerosols in the  $10 \mu\text{m}$  window will be very difficult because they scatter radiant energy very weakly. The aerosol layer for the example given here would scatter at most  $3 \times 10^{-5}$  of the energy from a lidar beam of infrared energy incident on the layer. Since the particles are about 2 orders of magnitude smaller than the wavelength of the  $10 \mu\text{m}$  energy, they scatter light according to the Rayleigh phase function. Hence the sulfate layer would scatter backwards per unit solid angle about  $5 \times 10^{-6}$  of the energy incident on it

(neglecting other interactions). An order of magnitude less energy is scattered in the most transparent band near to the strongly absorbing band. To take advantage of such contrast by a differential absorption technique, the numerous absorption lines of water vapor and trace gases have to be avoided.

Satellite mission requirements to measure only aerosol properties are not restrictive. Aerosols are distributed globally with lifetimes of about a week. Strong influxes of aerosols from fires or dust storms can be seen in satellite imagery 10,000 km from their sources. Satellite observations of the entire globe daily would seem to be sufficient.

The local time of the observations requires further study because of clouds. Cumulus clouds are less frequent in the morning over continents. In the absence of clouds, observations with a solar zenith angle of about  $60^\circ$  are favorable, either in the morning or afternoon. For the morning observations, the early morning fog has had time to dissipate. Also, the surface reflection of light from land is less at large solar zenith angle since the intensity of light reflected by the ground varies approximately as the cosine of the solar zenith angle, whereas the intensity of light scattered by the aerosols is not strongly dependent on the solar zenith angle. The major consideration, however, is the limitation caused by clouds, which is more pervasive than just the cumulus problem.

Satellite sensors for EOS will be able to measure the tropospheric aerosol number, surface area, and mass within the size range of 0.1 to 5  $\mu\text{m}$ . Clouds will limit the number of observations that can be made globally each day. Lidar can provide vertical profiles of aerosol physical parameters, but with low coverage. With the use of standard meteorological wind data, horizontal transport, convergence, and divergence can be calculated. The satellite data will be a poor source of information about aerosol chemical composition. The mass fraction of aerosols that absorb radiant energy and also the fraction that is condensed water can be estimated, however.

EOS will provide the basic instruments needed for tropospheric aerosol measurements. A Moderate Resolution Imaging Spectrometer (MODIS) is conceived to measure properties of the land, ocean, and atmosphere in 64 spectral bands, spanning the range between 0.4 and 1.0  $\mu\text{m}$ , plus bands at longer wavelengths. Except for choosing a band at a shorter wavelength of 0.35  $\mu\text{m}$ , measurements from more bands than are needed will be available for deriving aerosol optical properties since they change rather slowly with respect to wavelength and are well correlated. The spatial resolution of MODIS is 1 km at nadir, and it scans the entire Earth every 2 days. Such coverage would be augmented by measurements from other experiments. The aerosol properties can be estimated for the size range 0.1 to a few micrometers. New studies on deriving the aerosol properties from measurements in many bands and the accompanying algorithms are required.

There are currently no plans to continue SAGE II operations or to fly another occultation experiment in this time period. However, because of its relatively low cost, reliability, and capability to measure tropospheric aerosols in addition to stratospheric aerosols, consideration should be given to flying a modified version of SAGE to obtain multiwavelength extinction data for inferring aerosol optical properties and studying the interchange of constituents between the troposphere and stratosphere. Additional spectral channels could be added to obtain further size resolution of aerosol properties, as well as other gas species such as methane. Other improvements could include flying similar instruments on several platforms or tailoring the platform orbit to improve geographic coverage. Stellar and lunar

occultation measurements should also be considered, especially for EOS polar orbits where global coverage could be obtained almost daily by using several stars. The use of a SAGE device would also be important to correcting various passive remote sensors for aerosol effects.

Lidar observations of aerosols from space platforms such as EOS should provide the best vertical resolution data available, with good horizontal resolution ( $\approx 100$  km) along the satellite nadir-viewing track (subsattellite tracks will, however, be  $24^\circ$  longitude apart) and global coverage on a daily basis. Reasonably accurate (20 percent) aerosol backscatter coefficient profiles will be obtainable with 1 to 2 km vertical resolution, whereas the vertical resolution in mapping tropospheric layers such as Saharan dust or other continental plumes should be better than 100 m (Browell et al., 1985). Measurements of total columnar values (optical depth) by other sensors will complement the lidar data set by filling in gaps between subsattellite tracks.

Measurements of the chemical composition from satellites will be minimal or nonexistent. Composition may be inferred from the location of the aerosol source and properties that can be measured--such as absorption and size distribution. In addition, supplementary data from surface and aircraft observations will help to analyze the satellite observations for composition. The difficulties in measuring aerosol spectra that can identify the aerosol are discussed in this report.

One of the critical difficulties is lack of knowledge concerning the optical constants of aerosols, that is, their real and imaginary indices of refraction. Many aerosols consist of a mixture of substances. Their optical constants are not known and are difficult to measure. For continued progress in remote sensing of aerosol properties, their optical constants have to be measured.

#### PERIOD: 2000-2010

Two significant improvements in aerosol measurements are expected for this period. The first is higher lidar coverage spatially. Additional spectral bands will be available also. The second advance will be the measurement of winds from Doppler shifts in the light scattered from laser beams and also Doppler shifts in emission spectra from the limb of the atmosphere. In addition, studies are in progress to upgrade the geostationary satellites for this period by increasing the number of spectral bands and reducing the instantaneous field of view, thus reducing the cloud contamination problem. If the analyses of previous satellite data continue before this period, then scientists will be prepared to accept the challenges presented by new remote sensing observations.

#### REFERENCES

- Bohren, C. F. and D. R. Huffman, Absorption and Scattering of Light by Small Particles (John Wiley and Sons, New York), 1983.
- Browell, E. V., M. P. McCormick, S. Ismail and T. J. Swissler, Spaceborne Lidar System for Measurements of Atmospheric Water Vapor and Aerosols, AIAA-85-2091, 1985.
- Fraser, R. S. and Y. J. Kaufman, The Relative Importance of Aerosol Scattering and Absorption in Remote Sensing, IEEE Transact. Geosci. Remote Sens., GE-23, 5, 625-633, September 1985.

- Fraser, R. S. and Y. J. Kaufman, Calibration of Satellite Sensors After Launch, Appl. Optics, 25, 1177-1185, April 1986.
- Goetz, A. F. H., G. Vane, J. E. Solomon and B. N. Rock, Imaging Spectrometry for Earth Remote Sensing, Science, 228, 1147, 1985.
- Griggs, M., AVHRR Aerosol Ground Truth Experiment (Science Applications, Inc., San Diego, CA) 1984.
- Kaufman, Y. J. and R. S. Fraser, Remote Sensing of Aerosol Absorption, presented at International Radiation Symposium, Perugia, Italy, 1984.
- Kent, G. S. and M. P. McCormick, SAGE and SAM II Measurements of Global Stratospheric Aerosol Optical Depth and Mass Loading, J. Geophys. Res., 89, 5303-5314, 1984.
- McCormick, M. P., SAM II Measurements of the Polar Stratospheric Aerosol I, October 1978 to April 1979, NASA RP-1081, 1981.
- McCormick, M. P., Aerosol Measurements From Earth Orbiting Spacecraft, Adv. Space Res., 2, 73-86, 1983.
- McCormick, M. P., SAGE Aerosol Measurements, Volume I - February 21, 1979 to December 31, 1979, NASA RP-1144, 1985.
- McCormick, M. P., Patrick Hamill, T. J. Pepin, W. P. Chu, T. J. Swissler and L. R. McMaster, Satellite Studies of Stratospheric Aerosols, Bull. Am. Meteor. Soc., 60, 1038-1046, 1979.
- Martin, A. and F. R. Barber, Particulate Sulphate and Ozone in Rural Air: Preliminary Results From Three Sites in Central England, Atmos. Envir., 19, 1091, 1985.
- Mauldin, L. E., III, N. H. Zaun, M. P. McCormick, J. H. Guy and W. R. Vaughan, Stratospheric Aerosol and Gas Experiment II Instrument: A Functional Description, Opt. Eng., 24, 307-312, 1985.
- Pierson, W. R., W. W. Brachaczek, T. J. Truex, J. W. Butler and T. J. Korniski, Ambient Sulfate Measurements on Allegheny Mountain and the Question of Atmospheric Sulfate in the Northeastern United States, Annals New York Acad. Sci., 338, 145, 1980.
- Remsberg, E. E., Radiative Properties of Several Probable Constituents of Atmospheric Aerosols, The Infrared Handbook, pp. 4-70, 1978.
- Toon, O. B. and J. B. Pollack, A Global Model of Atmospheric Aerosols for Radiative Transfer Calculations, J. Appl. Meteor., 15, 225, 1976.
- Woodbury, Gerard and M. P. McCormick, Global Distributions of Cirrus Clouds Determined from SAGE Data, Geophys. Res. Lett., 10, 1180-1183, 1983.
- Yue, Glenn K., M. P. McCormick and W. P. Chu, Retrieval of Composition and Size Distribution of Stratospheric Aerosols with the SAGE II Satellite Experiment, J. Atmos. and Oceanic Tech., 3, 3, 371-380, September 1986.



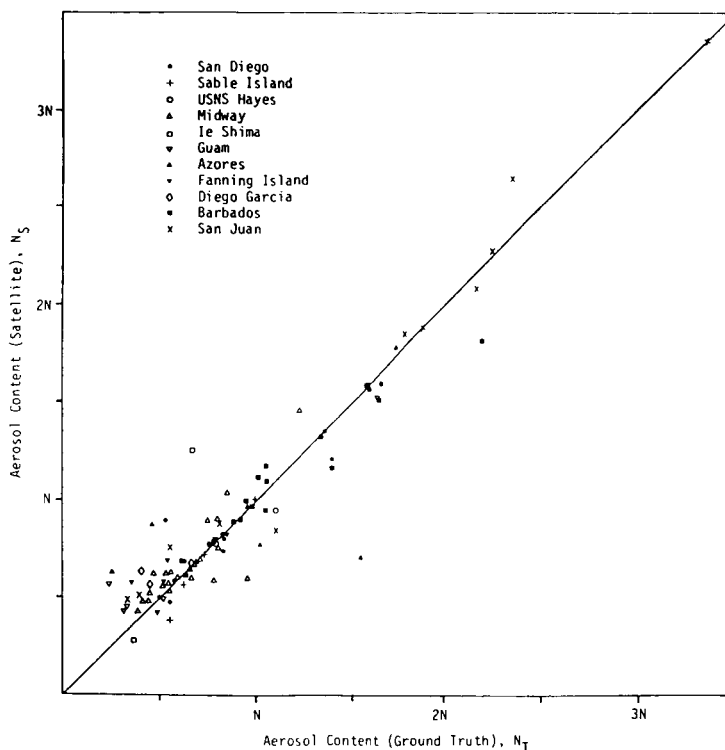


Figure 16.- Satellite measurements of aerosol optical thickness at  $\lambda$  500 nm by Griggs are compared with ground-based values. One N-unit is equivalent to 0.21 (Toon and Pollack, 1976).

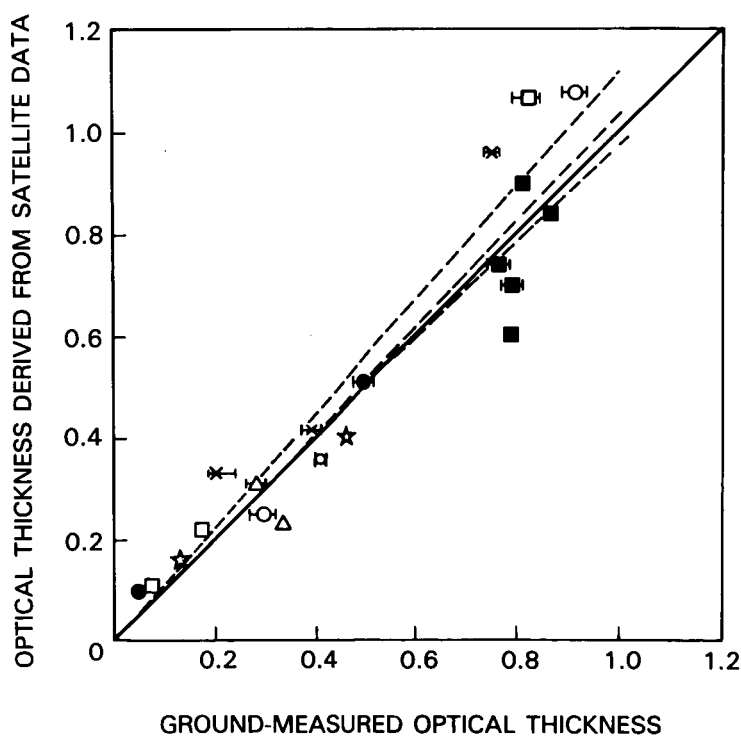


Figure 17.- Comparison of aerosol optical thicknesses measured from a geostationary satellite and the ground for  $\lambda$  610 nm. The three regression lines were calculated for 1000 sets of data by the bootstrap statistical method and include 34, 50, and 68 percent of those lines (Fraser and Kaufman, 1985a). Some of the ground-based measurements were made by Prospero (J. M. Prospero, private communication, 1982).

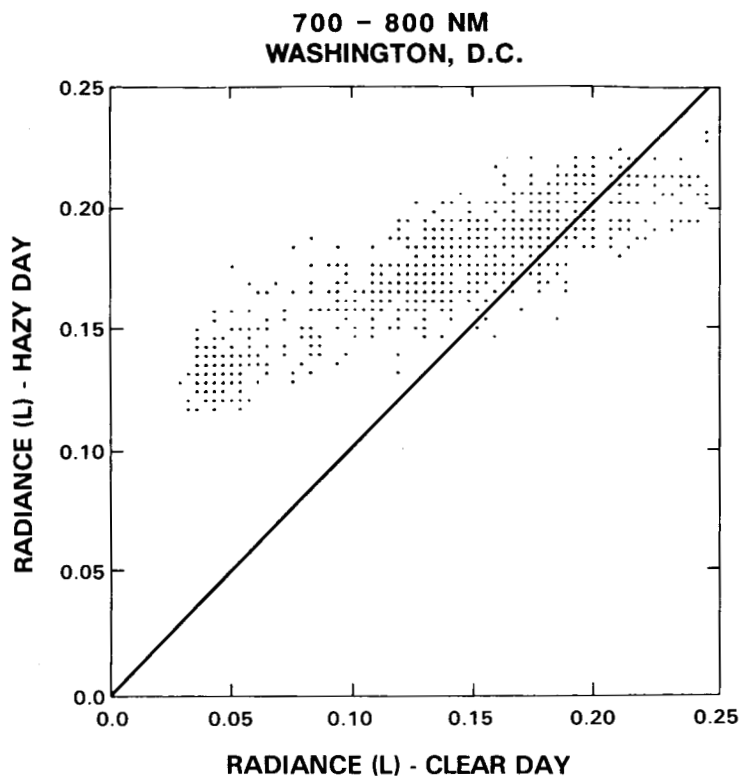


Figure 18.- The change in normalized radiance from a day of low aerosol content to one of high aerosol content in August 1982. The term  $L$  is the radiance in the Landsat 700-800 nm band;  $F_0$  is the extraterrestrial spectrum irradiance; and  $\mu_0$  is the cosine of the solar zenith angle (Kaufman and Fraser, 1984).

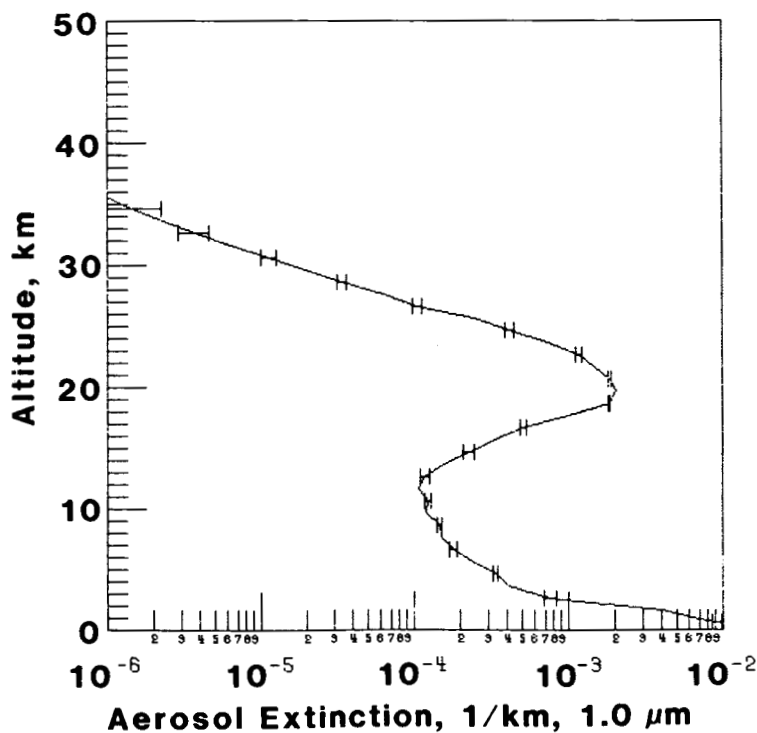


Figure 19.- Aerosol extinction ( $\text{km}^{-1}$ ) profile derived from SAGE II solar occultation measurements at  $\lambda$  1.02  $\mu\text{m}$ . One sigma error is shown as horizontal bars on the profile.